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THE TROPOSPHERIC OZONE MAXIMUM OVER THE TROPICAL SOUTH ATLANTIC OCEAN: A METEOROLOGICAL PERSPECTIVE FROM TRACE-A

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1. INTRODUCTION

NASA's Transport and Atmospheric Chemistry near the Equator--Atlantic (TRACE-A) experiment was conducted in 1992 to investigate the enhancement of tropospheric ozone over the south tropical Atlantic Ocean (Fishman et al., 1990). This anomaly was first identified from analyses of satellite data (see Figure 1) and was shown to be most pronounced during September and October. Subsequently, this feature was verified through a program of systematic ozonesonde releases from Ascension Island (8° S, 15° W) between 1990 and 1992 (Fishman et al., 1992). A comparison of the ozonesonde data with the climatological tropospheric ozone amounts derived from the satellite data is shown in Figure 2.

The existence of this feature generated several theories regarding its origin. One hypothesis suggested that its presence was the result of in situ formation from the oxidation of precursors emitted from widespread vegetation burning in Africa (Fishman et al., 1991). On the other hand, the role of transport processes was also shown to be important. Krishnamurti et al. (1993) suggested that transport from the stratosphere, as well as middle-latitude transport of emissions and the accompanying ozone formation resulting from vegetation burning in Brazil could contribute to the high levels of ozone over this region. With these questions posing as the motivation, TRACE-A was designed with the intent of using airborne, balloon-borne, satellite, ground-based, and conventional meteorological measurement methods to supply the information necessary to construct a viable model to explain the observations.

The purpose of the present study is to highlight some of the observations during TRACE-A and to provide a tentative explanation as to the origin of elevated ozone concentrations. To provide insight, we show a few examples of elevated ozone observations and explain them through the use of a trajectory model and measurements of other trace gases.

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2. THE TRACE-A FIELD EXPERIMENT

A series of 17 flights (143 flight hours) using the NASA DC-8 "Flying Laboratory" were carried out in September-October 1992. The flight paths are shown in Figure 3. Aboard the DC-8 were a suite of instruments capable of measuring all of the trace gases that are important for understanding how ozone is generated photochemically in the atmosphere, including oxides of nitrogen, hydrocarbons, nitric and organic acids, hydrogen peroxide, formaldehyde, acetaldehyde, oxygenated nitrates, as well as some longer lived trace gases such as carbon monoxide, methane, carbon dioxide and several halocarbons that could readily be used as tracers of airmass origin. Some flights were designed to characterize the local and regional composition of the air after being polluted by emissions from active fires in both South America and Africa. Additional flights were conducted to study chemical transformations of the air after it had undergone transport over the ocean for at least a day. Specific flights were also designed to investigate the large scale divergence over the south Atlantic since this is an area dominated by the persistence of strong subsidence throughout most of the troposphere. In addition to measuring wind, temperature and dew point on the DC-8, dropwinsondes were deployed to measure these fields below the airplane. Lastly, the use of Differential Absorption Lidar (DIAL) allowed us to determine the distributions of ozone and particulates both above and below the flight level of the DC-8 (e.g., see Browell et al., 1983).

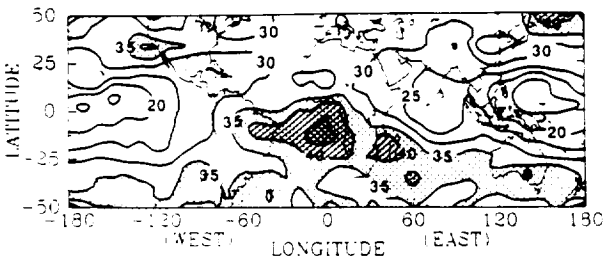

Figure 1. The climatological distribution of tropospheric ozone derived from satellite data is shown for October. The units of the contours are Dobson Units (DU) where 1 DU = 2.7 x 10^{16} molecules of ozone cm^{-2}. Values >35 DU are stippled, and those >40 DU are hatched and crosshatched (from Watson et al., 1990).

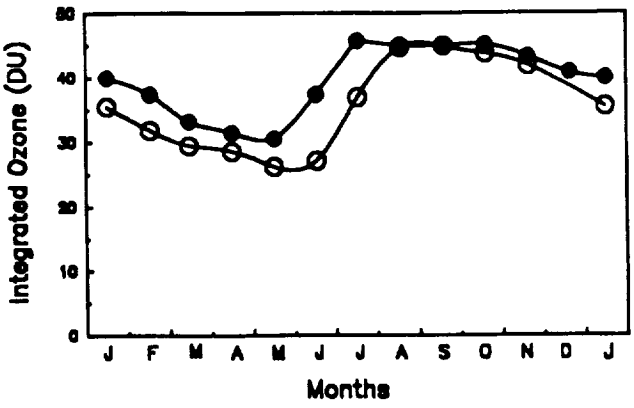

Figure 2. Comparison of the seasonal distributions of integrated tropospheric ozone at Ascension Island measured by ozonesondes (filled circles) and by satellites (open circles). All observations were put into monthly bins and then plotted on this figure using a 1-2-1 smoothing technique.

3. RESULTS

3.1 Case Study of 27 September 1992

The first example we present is one of the ozonesonde profiles obtained during the TRACE-A flight program. This particular profile is from 27 September 1992. The ozone concentration shown in Figure 4 is plotted in units of partial pressure and readily illustrates how most of the tropospheric ozone is confined to altitudes below ~6 km. Integrated throughout the troposphere (to a tropopause level of 120 mb), 65 DU of ozone are present, the highest integrated tropospheric ozone amount measured by any of the ozonesondes at Ascension Island. The temperature and dew point profiles show the presence of a strong subsidence inversion near 850 mb, a typical feature of nearly every profile from Ascension Island.

As shown in Figure 5, the prevailing synoptic situation is unremarkable. At 500 mb, the flow regime is dominated by an anticyclone centered near 17° S, 13° W. Another anticyclone is situated off the southwest coast of

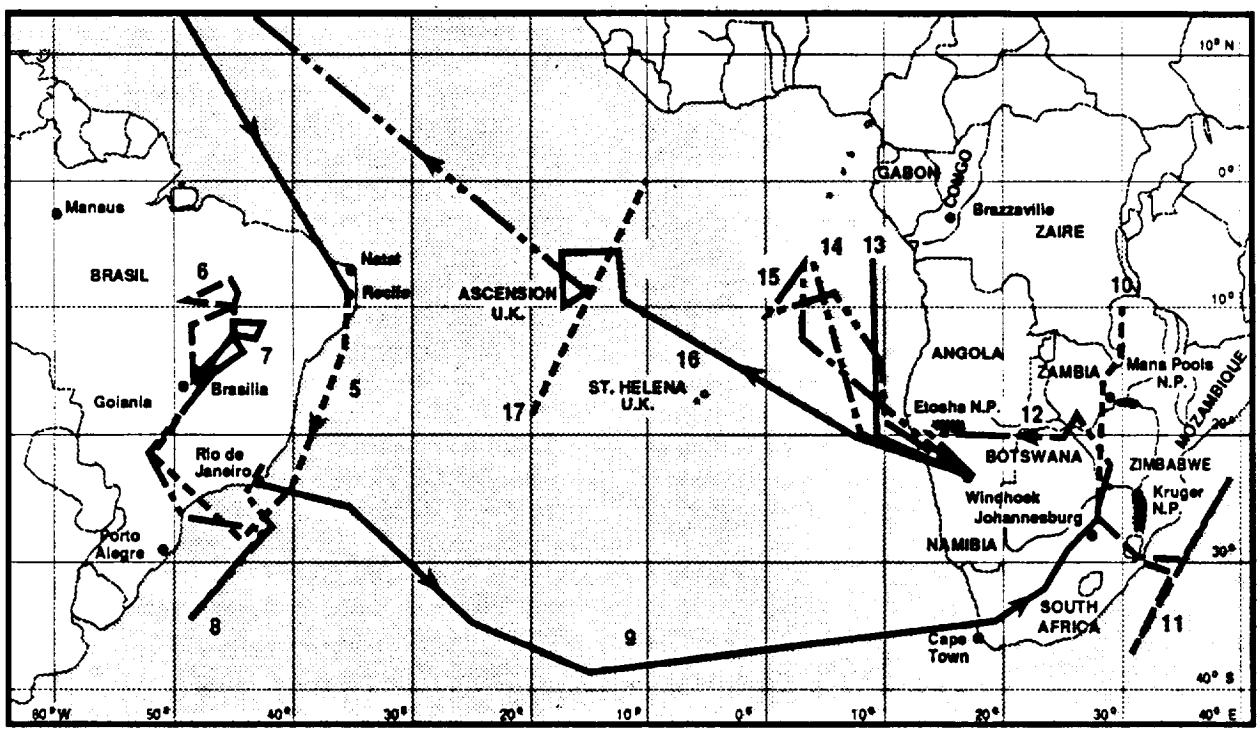

Figure 3. DC-8 flight paths during TRACE-A.

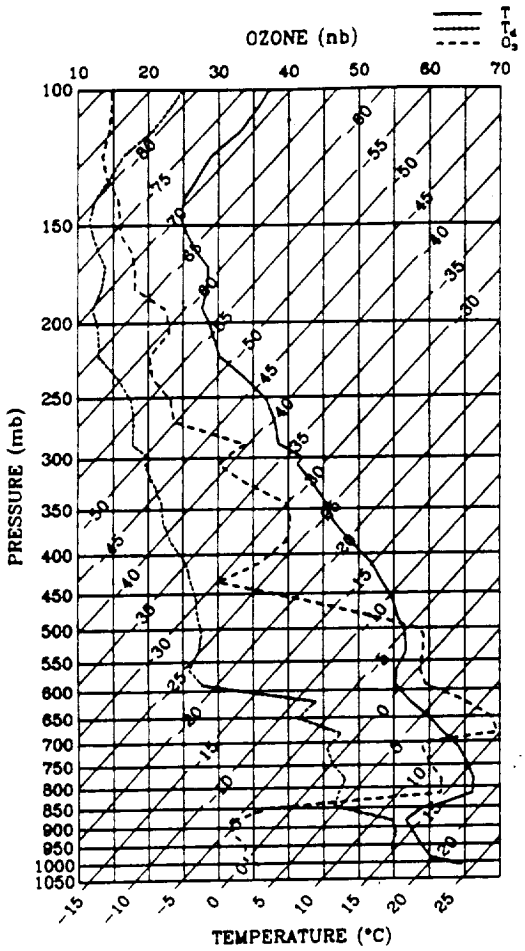

Figure 4. Ascension Island temperature, dew point, and ozone profiles for 27 September 1992.

Africa centered near 32° S, 11° E. At 850 mb and 700 mb (not shown here), the flow in the south Atlantic is generally characterized by weak easterly winds. At 300 mb (not shown here), winds are generally from the west or from the west-northwest at latitudes south of 10° S; closer to the equator, they are quite light and variable. None of these features at any of these altitudes is out of the ordinary for this region of the world.

A series of isentropic trajectory calculations (Haagenson and Shapiro, 1979) were performed to investigate some of the features of the ozone profile. The calculations were performed over a 7-day period for air parcels arriving at Ascension Island at 12 GMT on 27 September. The numbers in the box in the lower right corner of Figure 6 indicate the atmospheric pressure at which the parcels arrived at Ascension Island on the 27th. For altitudes below 526 mb, the air came from the east. Throughout this entire lower portion of the atmosphere, very high ozone concentrations were measured.

At altitudes that are characterized by lower ozone concentrations, the air appears to have been out to the west. Particularly intriguing is the trajectory calculated for the air at 438 mb. Although the air initially has an origin from the east, it appears to have become embedded in the anticyclonic flow shown in the 500-mb depiction in Figure 5. The air arriving at 224 mb, also of

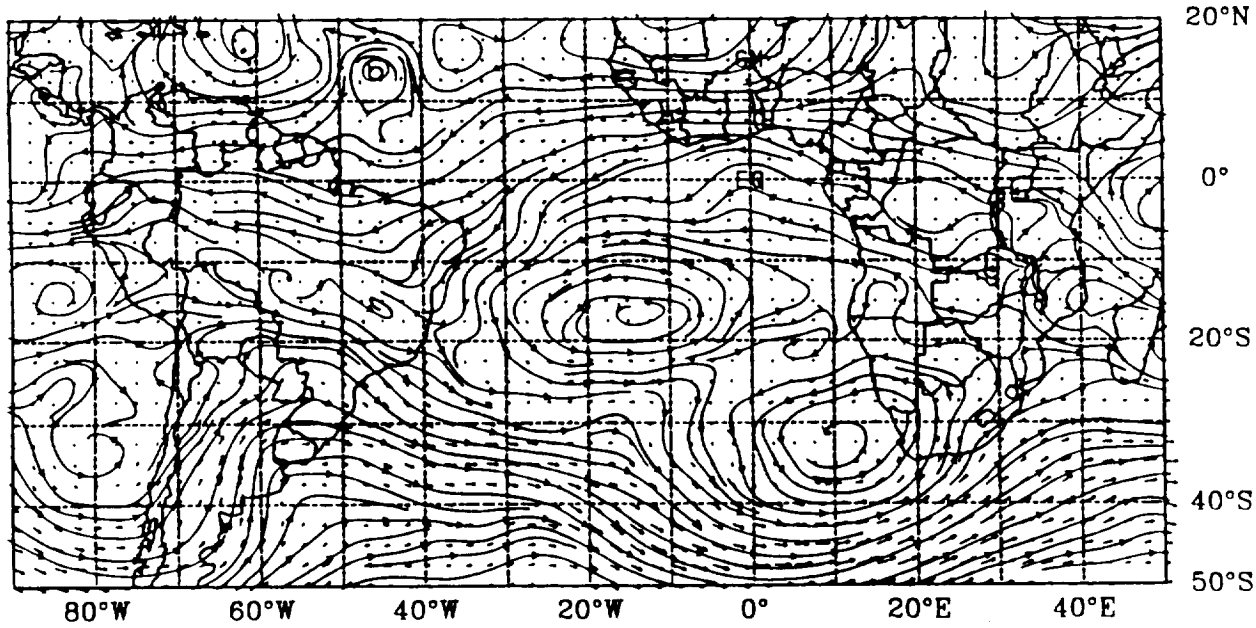

Figure 5. Streamline analysis at 500 mb for 27 September 1992.

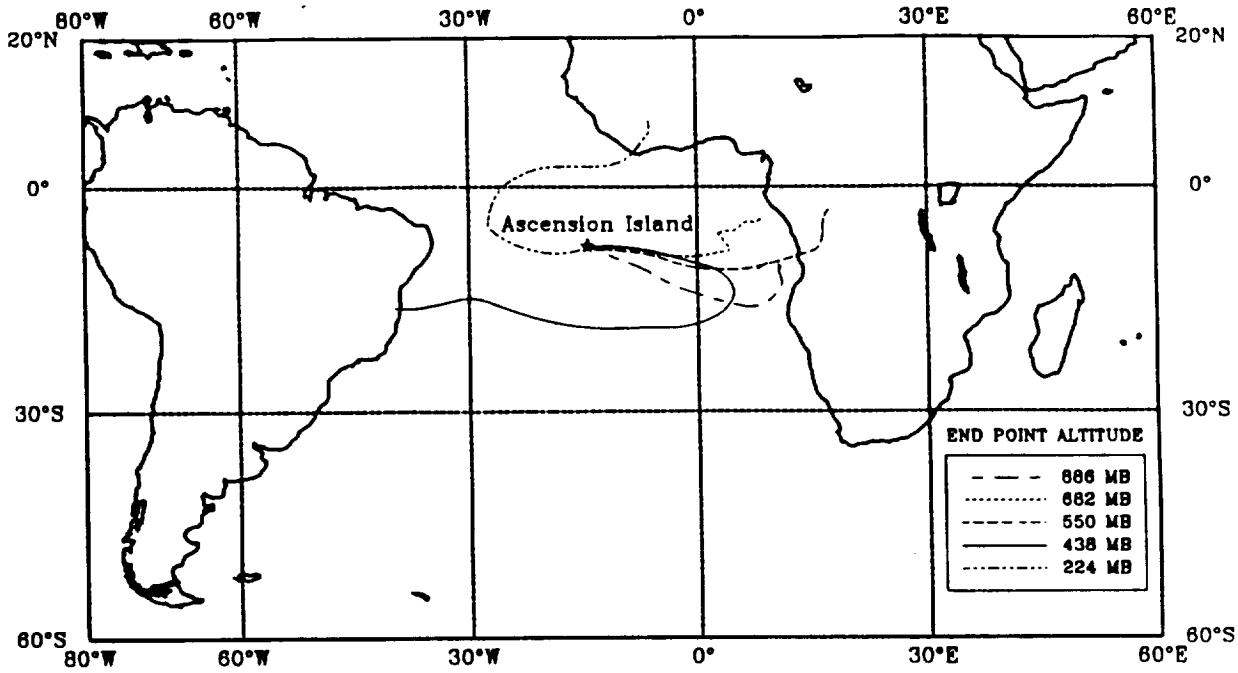

Figure 6. Seven-day isentropic analysis for air arriving at Ascension Island on 27 September 1992.

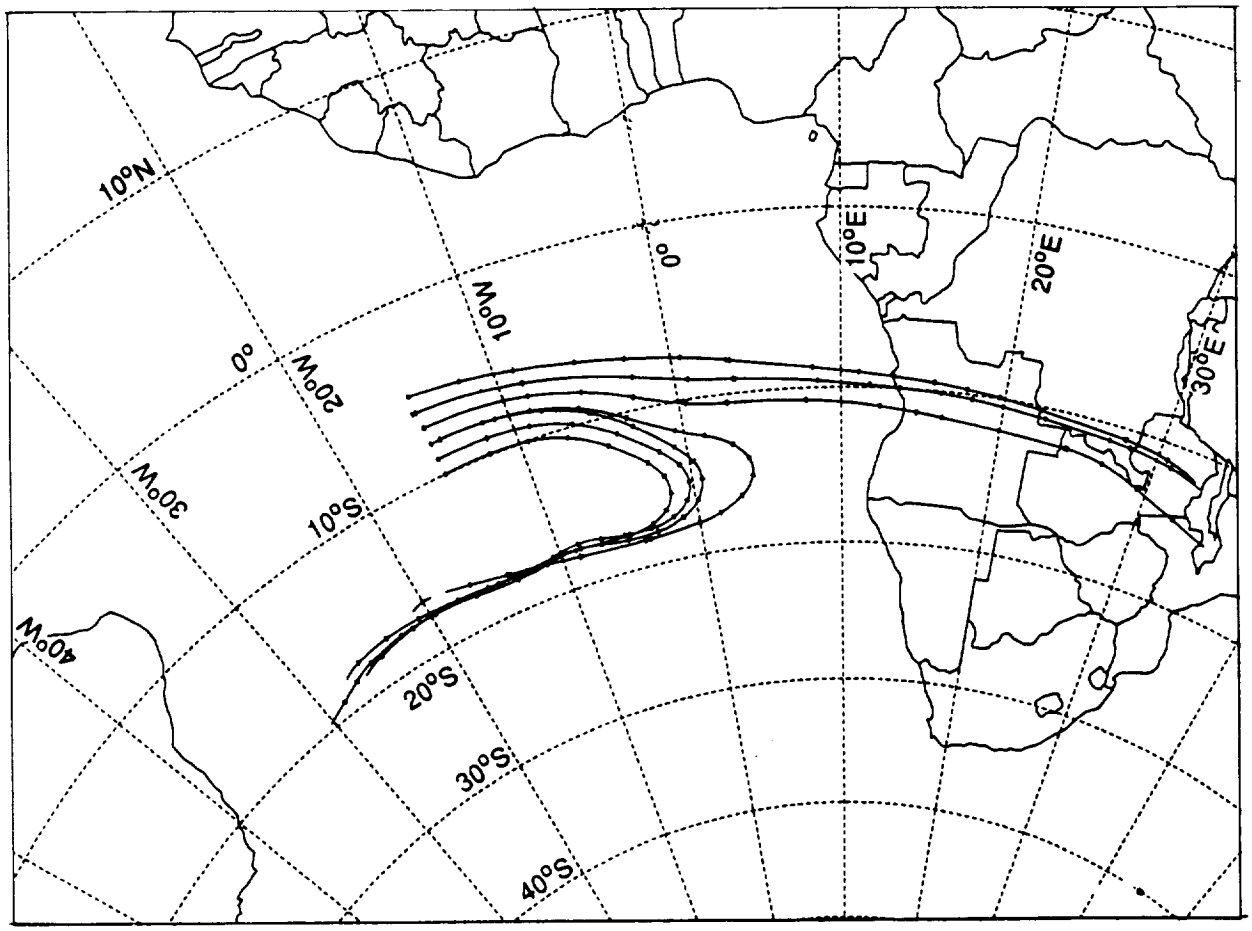

Figure 7. Isentropic trajectory calculations at 327° potential temperature along 15° W-meridian.

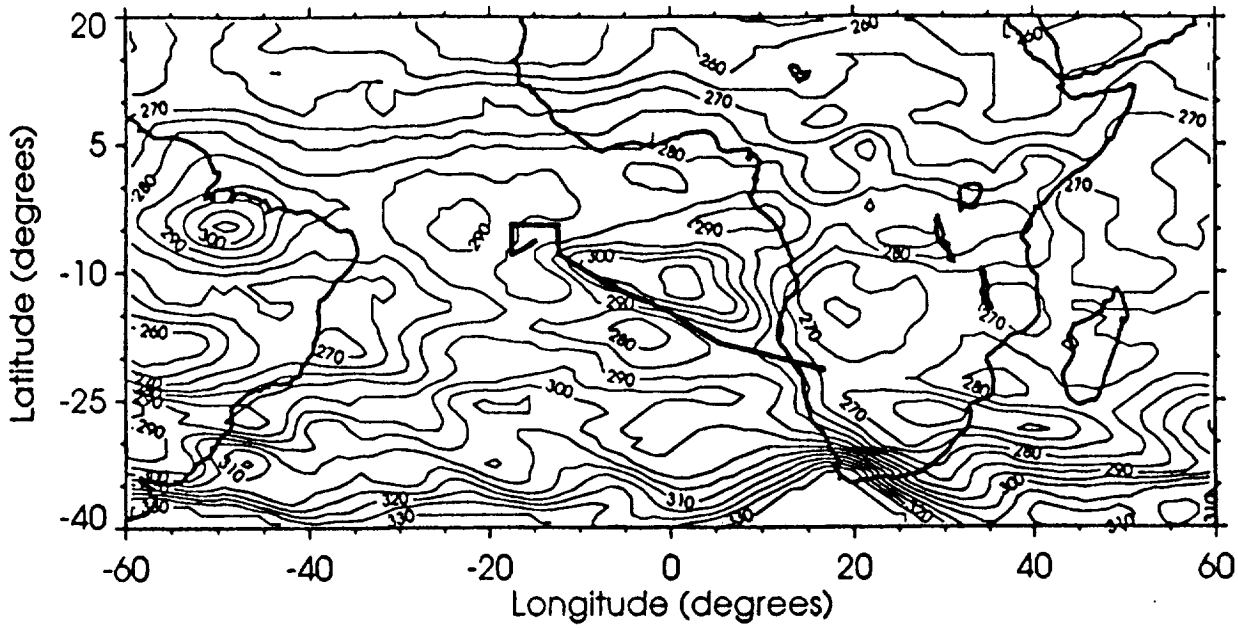

Figure 8. The flight track of the DC-8 is shown against the distribution of total ozone measured by the Total Ozone Mapping Spectrometer (TOMS) on 20 October 1992.

relatively lower ozone content, has been transported by the prevailing westerly flow in the upper troposphere, but appears to have been in northern Africa 7 days earlier.

This particular case study supports the hypothesis that ozone-rich air originates in Africa. We emphasize, however, that extrapolation of the results to form a climatology at the present time is not proper and that many more calculations need to be performed before any general conclusions can be drawn. One point that is illustrated by the above example is the complexity of the air flow in the region under investigation. An additional set of trajectory calculations is presented in Figure 7, which shows a set of six trajectories along the 15° W meridian for each degree of latitude between 5° S and 10° S. For the three northern points, the air had been over Africa, starting at approximately the same altitude as it ended on the 27th, for several days. The southern three trajectories, on the other hand, suggest that the air has been caught in the flow of the persistent anticyclone and had come from the west several days earlier. The calculations also suggest that the air has descended nearly 80 mb, a fact not surprising in light of the parcel's motion being strongly influenced by the anticyclone. The trajectories shown in Figure 7 also reiterate how sensitive they are to the position of the anticyclone. Even though it is a quasi-permanent feature of the south tropical Atlantic, the anticyclone does move and vary in intensity from one day to the next.

3.2 Case Study of 20 October 1992

This case study focuses on measurements obtained during the flight between Windhoek, Namibia (22° S, 17° E), and Ascension Island on 20 October. Figure 8 shows the flight path of the DC-8 on this day superimposed on the distribution of total ozone derived from an analysis of data from the TOMS instrument aboard the Nimbus-7 satellite. Although only 10-20% of the total ozone is in the troposphere, Fishman et al. (1990) suggested that measurements of total ozone at low latitudes can often be used as a means of identifying regions of enhanced ozone in the troposphere. As can be seen in Figure 8, the DC-8 flew close to an ozone maximum and definitely should have intercepted an ozone gradient between Windhoek and Ascension Island if indeed the TOMS depiction was representative of tropospheric ozone variability.

Plotted in Figure 9 are the ozone concentrations at flight level, the total ozone amounts derived from TOMS, and the integrated amount of ozone between 4-8 km determined from the DIAL measurements. During the transit to Ascension Island, flight altitude was 10.0 km between 0830 and 1045 UT and 11.2 km between 1045 and 1220 UT. The in situ ozone sensor measured ~100 ppbv (parts per billion, volume) between 0830 and 1010. A sudden drop from 96 ppbv to 72 ppbv occurred at 1010 with a steady increase to 92 ppbv until 1045, when the airplane shifted cruising altitude. At the new flight altitude of 11.2 km, ozone concentrations

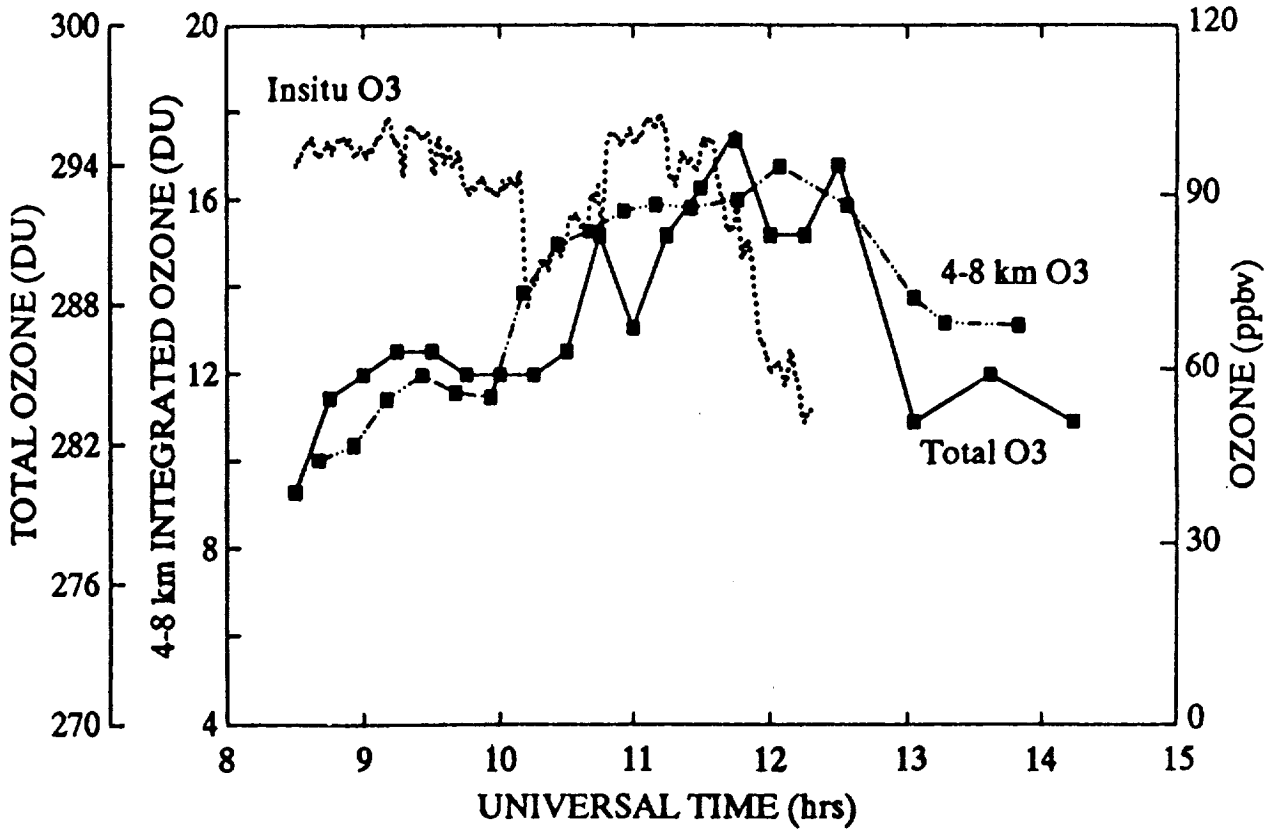

Figure 9. In situ ozone (dotted line), total ozone from TOMS (solid line) and 4-8 km integrated ozone obtained from DIAL (dash-dot line) along the flight path of DC-8, 20 October 1992, plotted as a function of time.

remained fairly constant at ~100 ppbv (with a range of 95-105 ppbv) until 1140. At that time ozone concentrations dropped significantly from values ~100 ppbv to ~50 ppbv by the time the plane started to descend at 1220 UT.

Relatively higher satellite total ozone is seen between 15° S and 8° S which was transected by the DC-8 between 1045 and 1220 GMT. Highest total ozone was 295 compared with 280 DU at the beginning of the flight. By the time of descent into Ascension Island, ~1420, total ozone was back down to 283 DU.

Integrated ozone below the airplane was obtained from the DIAL system. The integrated ozone between 4 and 8 km along the flight path is shown by the dash-dot line. This layer was chosen because DIAL data are available along the entire level-flight portion of the flight. Even though measurements are available for parts of the flight both above and below this layer, it is important that a uniform thickness is used for the entire part of the flight leg being analyzed so that comparisons between the DIAL integrated ozone data and the satellite data are consistent.

Relatively higher ozone was observed in the 4-8 km layer between 1015 and 1245 increasing to a maximum 16.8 DU near 11° S, 8° W from a value of 10.1 DU just off the Namibian coast. At the same time that the 4-8 km ozone started to increase at 1015, the in situ ozone at 10.0 km dropped sharply. The total ozone, on the other hand, remains relatively constant until 1045, suggesting that the increase in the lower troposphere is balanced by the decrease in the upper troposphere. By the time the DC-8 started to descend into Ascension Island, the integrated ozone in the 4-8 km layer was down to 13.1 DU; the TOMS total ozone also decreases.

Upon reaching a point at 9° S, 12° W, the plane flew a box pattern around Ascension Island and performed a ramp descent and ascent along the west wall of the box at the 17° W meridian. The remainder of this section will focus on the vertical distribution of ozone and other trace gases obtained during this portion of the flight and by an ozonesonde release at Ascension Island. Figure 10 compares the descent into Ascension Island starting at 1420 UT and ~100 km southwest of the runway with an ozonesonde profile obtained ~4 hr earlier. Some general features are captured by both profiles: generally high (>80 ppbv) concentrations between 5 and 10 km; an ozone minimum near 4 km; a layer above the trade wind inversion (1.5 - 4 km) with values generally 60-70 ppbv;

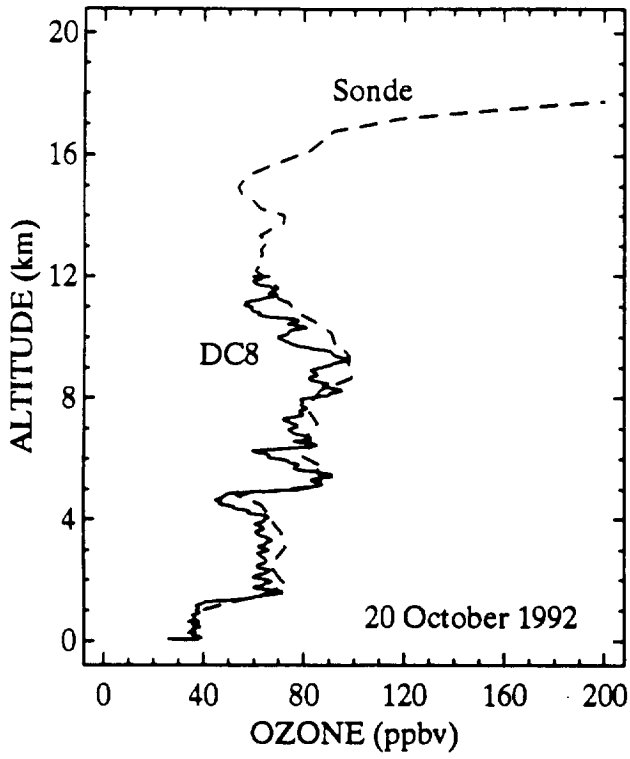

Figure 10. Comparison of ozonesonde and DC-8 ozone profile during descent into Ascension Island on 20 October 1992.

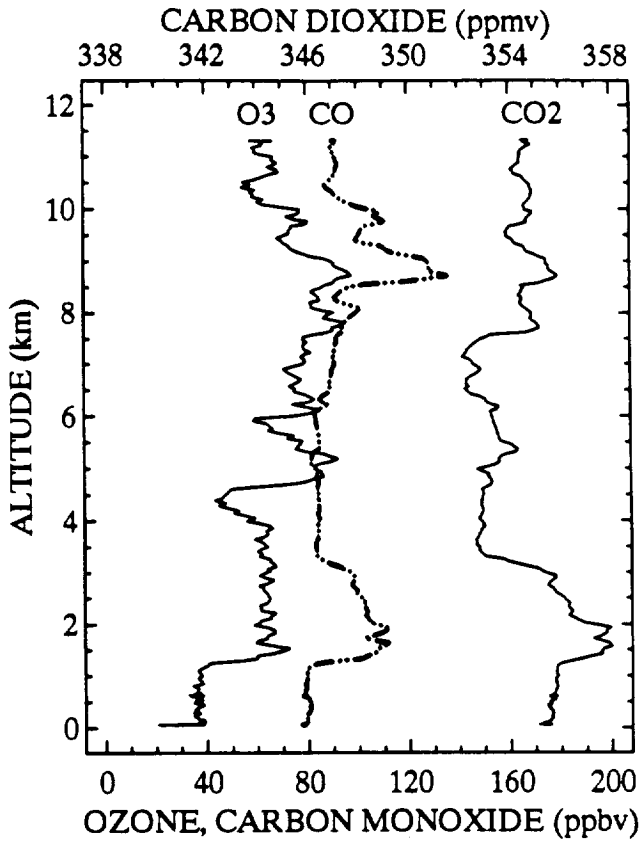

Figure 11. CO, CO_2, and O_3 profiles near Ascension Island for 20 October 1992.

and concentrations of 35-40 ppbv below the trade wind inversion. The ozonesonde shows a sharp increase in ozone concentration at 16.8 km, defining the tropopause. The agreement between the two profiles improves below 9 km, as the airplane gets closer to Ascension Island. Integrated from the surface to the tropopause, the profile contains 50 DU, a value slightly higher than the climatological average shown in Figure 2.

Figure 11 depicts the vertical structure of CO and CO_2 with ozone using fast-response sensors (>1 Hz) for each of the trace gases. These profiles show that the lamina of elevated ozone are clearly associated with higher concentrations of other trace gases of tropospheric origin. The most dominant ozone peak at 8.8 km is coincident with well-defined peaks of CO and CO_2. On the other hand, O_3 peaks at 5.3 and 7.8 km do not show coincident increases in CO, but do correspond with higher concentrations of CO_2. Such an enhancement would be consistent with the presence of an aged layer at these altitudes where most of the excess CO has been oxidized to ozone and carbon dioxide. Such an interpretation should be strengthened after a close examination of other trace gas species that are intermediate products and end products of the chemistry leading to ozone formation.

4. SUMMARY AND CONCLUSIONS

The presence of very high concentrations of ozone first identified by satellite over the remote south tropical Atlantic Ocean has been verified by measurements from aircraft and ozonesondes. These case studies show that elevated ozone concentrations are present at many levels in the troposphere; similar laminar structure is also observed in other trace gases of tropospheric origin supporting the hypothesis the excess ozone originates in the troposphere. Preliminary analysis of trajectories calculated for this study suggests that central and southern Africa is the region from which high ozone concentrations at Ascension Island originate. However, it must be emphasized that this analysis is based on only one such example, and that more general conclusions await further analysis. The suite of trace gases measured during TRACE-A will provide important information about the origin of this ozone maximum not available from the analysis of meteorological data alone.

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